

# Diffusion of the superconducting transition in HTSC

R. V. Vovk<sup>1</sup> · G. Ya. Khadzhai<sup>1</sup> · I. L. Goulatis<sup>1</sup> · S. N. Kamchatnaya<sup>1</sup> · A. Chroneos<sup>2,3</sup>

Received: 10 March 2017 / Accepted: 25 March 2017 / Published online: 3 April 2017  
© The Author(s) 2017. This article is an open access publication

**Abstract** In the present study it is shown that the transition to the superconducting state can be described within the framework of the phenomenological theory of diffused phase transitions, due to the structural material heterogeneity. The paraconductivity due to the appearance of fluctuating Cooper pairs at temperatures above  $T_c$ , demonstrates 2D–3D crossover, which also occurs in a certain temperature range, located above the superconducting transition interval.

It is well known [1–4], that in high-temperature superconductors (HTSC), the superconducting transition has a significant width in comparison to the classic superconducting transitions widths observed in pure metals. The broadening of the superconducting transition in HTSC is primarily associated with the inhomogeneous distribution of the labile oxygen concentration [5], which leads to phases with different oxygen deficiency in the sample, that is with different critical temperatures ( $T_c$ ).

At the same time, many phase transitions in real crystals exhibit a number of common features, regardless of their microscopic mechanism, such as the existence of pre-transition phenomena above the nominal critical transition

temperature, the formation of inhomogeneous structures during changes in temperature and/or by applying external fields the crystal [6–9]. Such transitions can be described by a general—for all kinds of transitions -phenomenological equation, describing the macroscopic volume increase of the new phase in the transition process [6].

The major common causes, that leads to diffused phase transitions (DPT) are: (1) large-scale heterogeneity of the physical system; (2) fluctuations of the physical quantities that characterize the state of the system in the phase transition (PT) region; (3) the impact of various internal and external fields; (4) surface or dimensional effects (PT in small particles, nanowires, thin films, etc.) [6].

The appearance of superconducting fluctuations higher than the critical temperature leads to predecessors of the superconducting phase, arising while the system is still in the normal phase, sometimes far away from  $T_c$  [10].

Obviously, for HTSC single crystals large-scale inhomogeneities of the distribution of the labile oxygen are relevant [4, 5], as well as superconducting fluctuations, causing paraconductivity (additional conductivity due to the fluctuation of the Cooper pairs, whose increase when approaching the superconducting transition leads to a reduction total resistance to zero) [10].

The phase state of the system with diffused phase transition is described by the inclusion function,  $L(T)$ , equal to the relative share of any of the phases [6]:

$$L(T) = \frac{1}{1 + \exp \frac{T_c - T}{w}} \quad (1)$$

This is a step-wise function, near the transition temperature,  $T_c$ . The parameter  $w$  characterizes the transition diffusion. The derivative of the inclusion function is portrayed by a curve with a final maximum:

✉ A. Chroneos  
alexander.chroneos@imperial.ac.uk

<sup>1</sup> V. N. Karazin Kharkiv National University, 4 Svobody sq., Kharkiv 61077, Ukraine

<sup>2</sup> Faculty of Engineering, Environment and Computing, Coventry University, Priory Street, Coventry CV1 5FB, UK

<sup>3</sup> Department of Materials, Imperial College London, London SW7 2AZ, UK

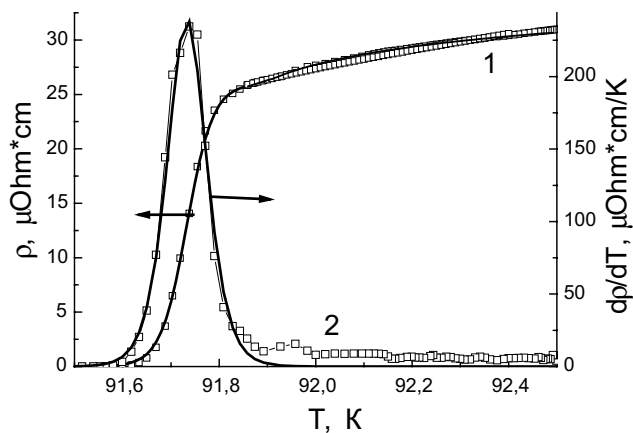
$$\frac{dL}{dT} = \frac{\frac{1}{w} \exp \frac{T_c - T}{w}}{(1 + \exp \frac{T_c - T}{w})^2} \quad (2)$$

We could note that the resistance measurements are carried out in a magnetic field of the measuring current and the superconducting transition, fixed on the electrical resistance is a type-I phase transition, close to type-II when the measuring current is small [6].

In the region of the superconducting transition, the resistivity of a homogeneous sample is proportional to the volume of the normal phase (which is formed due to various reasons), that is  $\rho \approx \rho_n L(T)$ ,  $d\rho/dT \approx \rho_n dL/dT$ .

The YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> single crystals were grown by the gold crucible solution–melt technology as described in detail in previous work [3]. Measurement of the electrical resistance of the samples was carried out by the standard four-point scheme using two pairs of silver contacts. The measurements were performed in the drift mode for two opposite directions of transport current to eliminate the impact of the false signal. The temperature was measured by a platinum thermistor, the voltage across the sample and the control resistance by V2-38 nano-voltmeters. The critical temperature was determined at the maximum point on the  $d\rho/dT$  curves near the superconducting transition. To reduce the oxygen content to the initial state the samples were annealed for a day in vacuum at 690 °C. After annealing, the crystals were rapidly cooled to room temperature in 2–3 min. Then, they were mounted in the measuring cell and cooled until liquid nitrogen temperature for 10–15 min. All measurements were performed at heated samples.

Figure 1 shows the  $\rho(T)$  and  $d\rho/dT$  dependencies of the optimally doped YBaCuO single crystal near the superconducting transition [11]. It can be seen that the width at half maximum of  $d\rho/dT$  is  $\approx 0.15$  K.



**Fig. 1** The superconducting transition in the optimally doped YBaCuO single crystal. 1 –  $\rho(T)$ , 2 –  $d\rho/dT$ . Points – experiment [11], lines – approximations:  $d\rho/dT$  by (2);  $\rho(T)$ —by (1) and (3)–(5)

Figure 1 also shows that the transition to the superconducting state occurs in two stages [12, 13]. The low temperature stage is connected directly to the superconducting transition and demonstrates  $d\rho/dT$  maximum and the high temperature stage is associated with the superconducting fluctuations, which are a pre-transition process. The  $d\rho/dT$  derivative at this stage is substantially less than in the low temperature stage.

The contribution of the superconducting fluctuations is obtained from the difference between the experimental conductivity and the conductivity extrapolated from high temperatures region ( $T \leq \theta$ ,  $\theta$  is the Debye temperature), where, obviously, there are no superconducting fluctuations.

We presume that the normal component of the electrical resistance of HTSC cuprates is determined by the scattering of charge carriers by phonons and defects. Subsequently the conductivity at the high temperature stage is expressed by the equation:

$$1/\rho(T) = 1/(\rho_0 + \rho_{ph}) + \Delta\sigma_{fluct}, \quad (3)$$

where  $\rho_0 = \text{const.}$ , is the residual resistivity;  $\rho_{ph}$  is the perfect resistivity: [14]

$$\rho_{ph} = A_3 \left( \frac{T}{\theta} \right)^3 \cdot \int_0^{\theta/T} \frac{x^3 dx}{(e^x - 1)(1 - e^{-x})} \quad (4)$$

and [10]

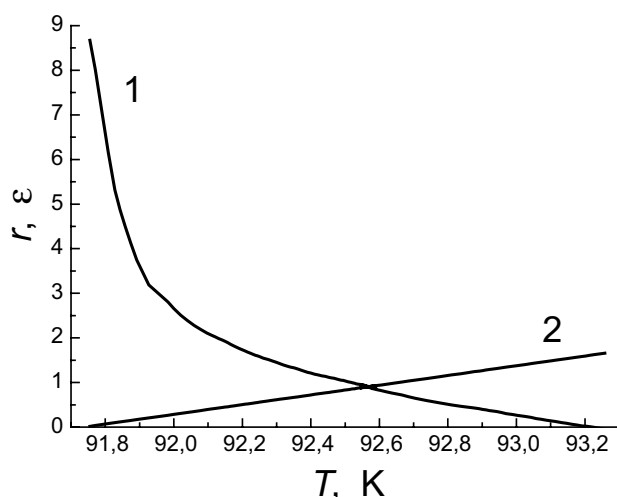
$$\Delta\sigma_{fluct} = \frac{e^2}{16d} \frac{1}{\sqrt{\varepsilon(\varepsilon + r)}}, \quad (5)$$

where  $\varepsilon = (T - T_c)/T_c$ ;  $r = 4\xi_c^2/d^2 = J^2/T$  is the Lawrence-Doniach anisotropy parameter;  $J$  is tunneling motion energy along the  $c$  axis [10, 15].

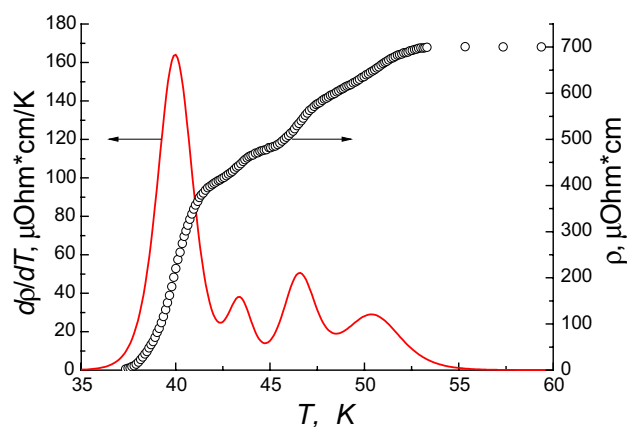
Since at  $\varepsilon < r$  the  $\Delta\sigma_{fluct} \sim \varepsilon^{-1/2}$ , which corresponds to the 3D case and in the opposite case at  $\varepsilon \gg r$  the  $\Delta\sigma_{fluct} \sim \varepsilon^{-1}$ , which corresponds to 2D case [10], then with a decrease in temperature (a decrease in  $\varepsilon$ ), a transition occurs from 2D to 3D motion of Cooper pairs, that is, a 2D–3D crossover. Naturally, the crossover occurs in a certain range of temperature changes. The temperature corresponding to  $\varepsilon = r$ , can be considered as the temperature of the 3D–2D crossover,  $T_{\text{cross}}$ . Wherein,  $\xi_c(\varepsilon_{\text{cr}}) \sim d$  [10].

This crossover is shown in Fig. 2, where the temperature dependences of  $\varepsilon$  and  $r$  are shown. It is seen that the transition from  $\varepsilon \gg r$  to  $\varepsilon < r$  occurs in the interval  $\approx 91$ – $94$  K, that is, the crossover is “diffused” along this interval. The intersection of  $\varepsilon(T)$  and  $r(T)$  occurs at  $T_{\text{cross}} \approx 92.6$  K. This point can be considered as the crossover temperature.

Figure 3 shows the  $\rho(T)$  and  $d\rho(T)/dT$  dependence for the oxygen substoichiometric sample (higher values of oxygen deficiency,  $\delta$ ) [16]. It can be seen that the entire transition to the superconducting state is expanded in the interval



**Fig. 2** Temperature dependencies of the reduced temperature,  $\varepsilon$ , and the anisotropy parameter,  $r$ . 1 –  $\varepsilon \times 100$ ; 2 –  $r \times 1000$



**Fig. 3** Plot of  $\rho(T)$  and  $d\rho(T)/dT$  for the underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  sample

$\approx 35$ – $55$  K and consists of distinct steps for  $\rho(T)$ , or distinct maxima for  $d\rho(T)/dT$ . This behavior can be connected with the presence of phases in the sample (probably meta-stable) with their  $\delta$  values and, respectively, their  $T_c$ .

Generally, these phases cannot be considered as connected in parallel or sequentially [17], that is, the resistance of such an inhomogeneous sample depends not only from the volume and the resistivity of the phase components, but also from the shape of the areas occupied by these phases. In particular, the vanishing of resistance at  $T < 37$  K can be caused not only by the fact that the entire sample is transferred to the superconducting state, but also by the fact that the phase which has the lowest  $T_c$  ( $\approx 40$  K), forms a superconducting percolation cluster, shunting the volume of the sample, still remaining in normal state.

In Fig. 3 is also observed that the width of maximum in each observed phase is  $\approx 2$ – $5$  K, which is much larger than the total width of the superconducting transition and the pre-transition process of the superconducting fluctuations appearance for monophasic sample (see Fig. 1). Such large widths of  $d\rho(T)/dT$  maxima, are apparently due to the existence of microdomains into the phase (similar to Kanzig fields in ferroelectrics [6, 18]) with different oxygen content and thus with different  $T_c$ .

Due to the different  $T_c$  within each phase, the superconducting regions coexist with regions where the fluctuation mode is implemented. Therefore, in a heterogeneous sample, to divide the pre-transition region (area of paraconductivity) and the actual superconducting transition area for each of the phases is not possible. For the whole inhomogeneous sample the allotment of the fluctuation region according to (3) – (4), as it was discussed in [16] carries an averaged, qualitative character.

Thus, in high-temperature superconductors the transition to the superconducting state occurs in a certain, sometimes quite wide, range of temperatures. This “diffusion” of the superconducting transition is due to the inhomogeneity of the material structure and to the presence of fluctuation Cooper pairs above  $T_c$ . In this case, a certain influence could have the specific mechanisms of quasiparticle scattering [19–23], due to the presence in the system of kinetic and structural anisotropy.

In the oxygen hypostoichiometric samples there exist macroscopic regions with different concentrations of labile oxygen, and, therefore, with different  $T_c$ , which generate steps in the  $\rho(T \approx T_c)$  dependencies, that corresponding to the superconducting transition in each of the phases. The superconducting transition for such each phase is blurred because of their micro-inhomogeneities and the presence of fluctuation Cooper pairs.

**Open Access** This article is distributed under the terms of the Creative Commons Attribution 4.0 International License (<http://creativecommons.org/licenses/by/4.0/>), which permits unrestricted use, distribution, and reproduction in any medium, provided you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

## References

1. D.M. Ginsberg ed. *Physical Properties High Temperature Superconductors I*. (Word Scientific, Singapore, 1989)
2. A.L. Solovyov, L.V. Omelchenko, V.B. Stepanov, R.V. Vovk, H.-U. Habermeyer, P. Przyslupski, K. Rogacki, *Phys. Rev. B* **94**, 224505 (2016)
3. A.V. Bondarenko, V.A. Shklovskij, R.V. Vovk, M.A. Obolenskii, A.A. Prodan, *Low. Temp. Phys* **23**, 962–967 (1997)

4. R. Menegotto Costa, F.T. Dias, P. Pureur, X. Obradors, *Physica C* **495**, 202 (2013)
5. R.V. Vovk, N.R. Vovk, O.V. Dobrovolskiy, *J. Low Temp. Phys* **175**, 614–630 (2014)
6. B. N. Rolov, V. E. Jurkiewicz, *The Physics of Diffuse Phase Transitions*. (Publishing house of the RSU, Rostov n / D. 1983), p. 319
7. G. A. Maligin, *Solid State Phys.* **35**, 127–137 (1993)
8. D.D. Balla, A.V. Bondarenko, R.V. Vovk, M.A. Obolenskii, A.A. Prodan, *Low. Temp. Phys* **23**, 777–781 (1997)
9. R.V. Vovk, G. Ya. Khadzhai, O.V. Dobrovolskiy, Z.F. Nazyrov, I.L. Goulatis, *Mater. Res. Exp.* **1**, 026303 (2014).
10. A. Larkin, A. Varlamov, *Theory of Fluctuations in Superconductors*. (Oxford University Press, USA, 2009)
11. R.V. Vovk, G.Ya.. Khadzhai, I.L. Goulatis, A. Chroneos, *Physica B* **436**, 88 (2014)
12. P. Pureur, R. Menegotto Costa, P. Rodrigues Jr., J. Schaf, J.V. Kunzler, *Phys. Rev. B* **47**, 11420 (1993)
13. C.J. Lobb, *Phys. Rev. B* **36**, 3930 (1987)
14. L. Colquitt, *J. Appl. Phys* **36**, 2454 (1965)
15. W.E. Lawrence, S. Doniach, *Proceedings of the Twelfth International Conference on Low Temperature Physics*, edited by E. Kanda, Academic Press of Japan, Kyoto, p. 361 (1971).
16. G.Ya.. Khadzhai, R.V. Vovk, Z.F. Nazyrov, *Low. Temp. Phys* **41**, 874 (2015)
17. P.L. Rossiter, *The Electrical Resistivity of Metal and Alloys*. (Cambridge University Press, Cambridge, 1987)
18. W. Kanzig, *Ferroelectrics and Antiferroelectrics*. Academic Press Inc., U.S., 1964, 197 & #1088.
19. R. V. Vovk, G. Ya. Khadzhai, and O. V. Dobrovolskiy, *Appl. Phys. A* **117**, 997–1002 (2014)
20. V.N. Golovach, M.E. Portnoi, *Phys. Rev. B* **74**, 085321 (2006)
21. I.N. Adamenko, K.E. Nemchenko, V.I. Tsyganok, A.I. Chervanov, *Low. Temp. Phys* **20**, 498 (1994)
22. A.L. Solovjov, L.V. Omelchenko, A.V. Terekhov, K. Rogacki, R.V. Vovk, E.P. Khlybov, and A. Chroneos, *Mater. Res. Express* **3**, 076001 (2016)
23. P.G. Curran, V.V. Khotkevych, S.J. Bending, A.S. Gibbs, S.L. Lee, A.P. Mackenzie, *Phys. Rev. B* **84**, 104507 (2011)